Global geochemical mapping and sediment-associated flux of major world rivers

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A global geochemical mapping project based on sampling of overbank sediments from rivers was established by the International Commission on Continental Erosion, ICCE, in 2001. Overbank sediments were introduced as a sample medium in geochemical mapping in 1989, and a number of papers on the geochemistry of overbank sediments have since been published. The present paper reviews main aspects of this literature, arriving at the following conclusions: Depth-integrated samples of overbank sediments reflect the composition of many current and past sediment sources upstream of the sampling point, contrary to active stream sediments which normally are recent deposits derived from a more restricted number of presently active sediment sources. Mapping the composition of recent and pre-industrial overbank sediments can therefore be used (1) in a characterisation of the present state of pollution, and (2) as a regional prospecting tool in natural as well as polluted environments. Human interference with rivers and the predicted climate change will affect the global sediment flux to the oceans.

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Introduction

Available data concerning the spatial heterogeneity of the chemical composition of the Earth's surface and data concerning the flux of persistent organic pollutants and metals from land to the marine environment are incomplete. Overbank sediments have turned out to be an important medium for the construction of maps of geochemical elements in large regions (Ottesen et al. 1989). Widely spaced, global geochemical sampling of deposited sediments is crucial because it provides a practical way to relatively quickly obtain a consistent overview of the contemporary global distribution of elements (natural and anthropogenic) on the Earth's surface. Such data can be used for environmental and related human health studies and mineral exploration.

The traditional method in geochemical mapping has been to sample sediments from the bottom of small streams. It has been assumed that the composition of active sediments in the stream channel represents the geochemistry of the catchment area upstream of the sample site. However, the sediments in stream channels are often derived from sources of limited extent and may change with time (Ottesen et al. 1989). The assumption that active stream sediments may characterise the composition of whole drainage areas can, therefore, be questioned.

Ottesen et al. (1989) suggested that overbank sediments could be a more representative type of sample and demonstrated that this type of sediment was usable also in very large drainage basins. Overbank sediments are deposited by large-magnitude floods during conditions when a number of sediment sources are active and thus integrate material from a large area. They are available along rivers with variable water discharge. Older sediment deposits are often preserved, providing opportunities to detect natural and anthropogenic chemical signals from the past. Many papers have subsequently been published about the use of overbank sediment as a geochemical sampling medium (e.g., Leenaers 1989, Lewis and Macklin 1989, Edén and Björklund 1994, Macklin et al. 1994, Ridgway et al. 1995, Bølviken et al. 1996, 2004, Langedal 1996, 1997a, De Vos et al. 1996, Gäbler 1997, Swennen and Van der Sluys 1998, 2002, Swennen et al. 1998, Walling and He 1998, Xie and Hangxin 2001, Caritat et al. 2005). Based on the use of overbank sediments, Xie and Hangxin (2001) mapped China (9.5 million km²) with 529 sample localities. The authors concluded that a global reconnaissance study should use overbank sediments as a sample medium.

A river is a dynamic system where continuous erosion and sedimentation give rise to a redistribution of sediments and sediment-associated chemical elements. The use of overbank sediments is also important when the sedimentary dynamics of a river system have to be taken into account. Changes in the hydrological regime, or man-made changes in sediment sources or transport conditions, may thus affect the distribution of chemical elements within the drainage basin and the sediment delivery to the oceans. A large part of the sediment flux to the oceans is delivered by a relatively small number of large rivers. A record of the changes in sediment supply due to natural or manmade changes in the sediment budget and the particle-associated flux of the world's major rivers, is thus of great importance.

In January 2001, the project 'Global geochemical mapping and sediment-associated flux of major world rivers' was established by the International Association of Hydrological Sciences/International Commission on Continental Erosion (IAHS/ICCE). It is the aim of this project to carry out a worldwide sampling programme based on overbank sediments to obtain a global geochemical map.

Formation of overbank sediments

Overbank sediments (also called alluvial soils, floodplain sediments or levee sediments) occur along rivers with variable water discharge. During floods, temporarily enhanced discharge may exceed the capacity of the channel (Figure 1a). Material in suspension will be transported and deposited onto floodplains and levees (Figure 1b). In most rivers, this process has taken place many times in the past.

Overbank deposits therefore consist of successive, nearly horizontal strata of young sediments overlying older sediments. A vertical section through such a deposit reflects the history of sedimentation back in time.

During floods, the great quantity of water activates many sediment sources in the drainage area, and the material in suspension will reflect the composition of these and earlier developed sources. This is why overbank sediments represent large parts of—or even complete—catchments.

In some overbank deposits, the stratigraphy may be complex due to redeposition of material eroded from earlier-formed upstream deposits. Young sediments may then be intermixed with older sediments.

Properties of overbank sediments

Since overbank sediments normally consist of individual horizontal strata formed at different times, the variations in chemical composition and the corresponding sampling error will be greater in the vertical plane than in the horizontal direction.

In principle, vertical trends in the chemical composition of overbank sediments have two origins, namely variations in the composition of the original source material and alteration caused by the secondary migration of substances after deposition. Secondary migration is due to factors such as climate, pH, reduction/oxidation conditions, the amount and type of organic material, biological activity and time. In many climates, soil formation processes may need hundreds of years in order to develop significant vertical patterns. For overbank sediments, the available time interval is normally more restricted, as new layers of sediment are deposited on top of the older ones. Problems of Figure 1. (a) Major flood in eastern Norway, and (b) a profile in the floodplain.



vertical migration are therefore expected to be less in overbank sediments than in other soils.

In Belgium, the Netherlands, Luxembourg and parts of Germany, 34 overbank sediment profiles located along the banks of meandering rivers have been studied (De Vos et al. 1996, Swennen and Van der Sluys 1998, 2002, Swennen et al. 1998). In 30 of these, pre-industrial sequences could be detected below polluted surface overbank deposits. Samples were collected at depth intervals of 10 cm and analysed for main and trace elements.

Three main groups of vertical distribution pattern were distinguished in the sections: (1) either low or high metal concentrations throughout the profile, thought to reflect high or low natural metal contents in the catchments, (2) no variations in grain size or lithology, but a gradual increase in heavy-metal concentrations towards the top of the profile, presumably caused by airborne pollution, and (3) abrupt changes in metal concentrations at certain depths and a corresponding change in lithology. These patterns are interpreted as being an effect of man-made discharges into the catchments and subsequent fluvial dispersion of particle-bound pollution.

In a Norwegian study (Langedal 1996, 1997a, b, c), overbank-sediment profiles were sampled from the Knabeåna– Kvina drainage basin, which is influenced by Cu- and Mo-rich tailings from the now closed Knaben molybdenum mine. Along the rivers, pre-industrial overbank sediments were detected below the present inundation level in the bottom sections of 14 out of 18 profiles. The four atypical profiles are situated where lateral river migration has had an impact on the sedimentary environment, or where minor river regulations and influx of tailings have altered the original peat bog and lacustrine environments into floodplains. Most of the Knabeåna–Kvina profiles show high Cu and Mo contents in the upper part, while concentrations in the bottom section approach a lower, probably natural level similar to the recent natural sediments that overlie the present inundation zone of polluted sediments.

Edén and Björklund (1994) suspected downward percolation of long-range atmospheric pollution to be the cause of high Pb concentrations in the lower part of the overbank-sediment profiles in northern Europe. Acid rain and low buffer capacity in the sediments may have contributed to the migration. However, Ottesen et al. (2000) questioned this interpretation, claiming that the Pb enrichments are natural, on the basis of data published by Taylor and Heier (1958), which reveal high Pb contents in K-feldspar in this region.

In English and Welsh basins with old Pb/Zn-mines, the vertical distributions of Pb and Zn in dated overbank-sediment sequences were found to be closely related to the mining history, suggesting that no significant vertical migration of these metals had taken place after the sediments had been deposited (Macklin et al. 1994). Similarly, along the Rio Guanajuato and Rio Puerco rivers, Mexico, no vertical migration was seen for the elements As, Cr, Cu, Pb, Sn, and Zn (Ridgway et al. 1995).

In a study of 49 selected floodplains, Edén and Björklund (1994) found that the lateral variations within floodplains were insignificant in comparison with the between-floodplain variation (Table 1). Similar results were obtained by Chekushin et al. (1993) for the chemical composition of duplicate overbanksediment samples from the border area of Finland, Norway and Russia.

Langedal (1997b) found that in floodplain surface sediments (0–25 cm depth) of the polluted Knabeåna river, the highest Cu and Mo concentrations are in samples proximal to the river or in floodplain depressions. Enrichment of metals in these parts of the floodplains may be an effect of differences in the timing of the sediment transport pulse, and the timing of floodplain inundation (Bradley 1984). In the proximal areas and the depressions, the suspended-sediment transport rates are often highest during the rising and peak stages. These are the first to be inundated and receive the largest load of particle-bound metals. Similar results were also found along the Geul river in Belgium by Leenaers (1989) and Swennen et al. (1994).

In both small and large catchments, the sampling error for natural overbank sediments within a floodplain is small

Table 1. Analysis of variance of the content of aqua regia-soluble chemical elements in widely spaced duplicate samples of overbank sediments taken at depth and near the surface in a 23,000 km² area in northern Europe. (From Edén and Björklund 1994).

Element	(1) %	(2) F	(3) F	(4) F
Al	2.2	14.5	6.1	4.2
Ва	6.0	14.7	5.6	5.4
Ca	1.8	73.0	15.3	15.3
Co	10.0	12.0	5.8	7.8
Cr	4.7	48.0	7.1	8.3
Cu	10.7	34.7	5.6	5.3
Fe	25.5	13.0	5.1	4.4
К	3.8	33.3	10.5	10.8
La	4.3	34.0	9.3	6.1
Mg	1.8	66.0	8.6	10.3
Mn	6.2	14.1	3.7	6.0
Na	7.3	4.2	5.6	4.6
Ni	10.1	26.0	7.3	7.9
Р	4.0	25.0	6.3	8.9
Pb	21.7	5.5	3.9	4.1
Sr	5.0	47.2	8.2	8.3
Th	35.8	10.7	3.9	5.6
Ti	2.2	33.3	6.7	6.9
V	4.5	7.0	6.0	5.0
Zn	5.0	30.0	6.2	7.7
Critical F value at p=0.05		1.7	1.4	1.4
Number of pairs	36	36	116	116

(1)–(3) Samples at depth. (1) Combined sampling and analytical error. (2) Ratio between total variance and combined sampling and analytical variance. (3) Ratio of between-site variance and within-site lateral variance.
(4) Ratio of between-site variance and within-site vertical variance.

in relation to variation between floodplains. This conclusion appears to be valid in most regions of the world for genuinely natural deposits and in situations where pristine sediments at depth are covered by polluted surface sediments.

Sampling of older terraces is appropriate in order to obtain pre-industrial material. Such sampling should be done above the present inundation zone to avoid material draped during recent floods. Along laterally stable river reaches, sampling in the bottom sections of the sediment profiles is adequate (Langedal 1997a, b). Sampling along meandering reaches, as suggested by Bogen et al. (1992), may also be a possibility if lateral migration is slow.

Pollution of overbank sediments may be of two types: (1) mine waste and other anthropogenic material may enter the stream from local sources and then be transported downstream, and (2) airborne contaminants originating from distant sources may reach the catchments (Langedal and Ottesen 1998). Situation (1) is often recognisable since the sources may be easily identified, but situation (2) can be more difficult to detect.

The selection of appropriate locations for sampling overbank sediments should be undertaken only by personnel trained in sedimentology. If this prerequisite is fulfilled, subsequent highquality chemical analysis of the samples will produce reliable data for most chemical elements.

Regional distribution of chemical elements in overbank sediments

Ottesen et al. (2000) published a geochemical atlas of Norway based on 700 floodplain sampling sites distributed across the country (300,000 km²). Each site represents a drainage area of 60–600 km². A vertical section through the sediments was cut with a spade, and a composite bulk sample 50–100 cm long was taken from the section excluding the upper 5–10 cm. After drying, the samples were sieved and the < 0.062 mm fraction analysed for the total contents (XRF) of 30 elements and determination of 29 elements in a nitric acid extract. Most elements show systematic patterns with great contrast. In some cases, these patterns agree with those of known geological structures, in others they indicate previously unknown structures.

Xiachu and Mingkai (1995) carried out a reconnaissance survey in a part (170,000 km²) of the Jiangxi Province of southern China in order to develop techniques for implementing ultra-low density sampling of overbank sediments for global geochemical mapping. Sampling sites (1 site per 1800 km²) were picked at the apexes of 94 drainage basins of between 100 and 800 km². It was concluded that floodplains with catchment areas of 100 to 800 km² do indeed provide suitable sampling stations for global geochemical mapping based on overbank sediments. Widelyspaced sampling of lower-layer overbank sediments was seen as a fast and cost-effective method for identifying geochemical provinces.

Xie and Hangxin (2001) collected 529 floodplain samples across the Republic of China (9,600,000 km²), each sample representing a drainage basin of between 1000 and 6000 km². It was concluded that the geochemical data generated from the widely-spaced sampling were strikingly similar to those generated by China's Regional Geochemical National Reconnaissance Program, which was based on more than 1 million samples of active stream sediments.

As a contribution to the Geochemical Atlas of Europe, 747 floodplain samples from all over Europe were collected and analysed for both major and trace elements (Figure 2, Salminen et al. 2005). Caritat et al. (2005) published a geochemical survey based on the contents of a wide range of chemical elements in overbank sediments from parts of Australia. All these projects demonstrate that the use of overbank-sediment samples in regional geochemical mapping is a very cost-effective method.

Flux of major world rivers

A comparison of the annual sediment transport in about 20 major rivers reveals that there is no direct relation between the volume of the sediment load and the size of the river basins (Table 2). However, it is indicated that a small number of rivers deliver a large share of the total sediment flux to the oceans. The combined sediment load of the 20 rivers constitutes nearly half of the estimated total global load of 13,500 x 10^6 t yr⁻¹ that was given by Milliman and Meade (1983). Panin (2004) discussed the current estimates of global loads proposed over the last two decades and found that they all fall within the range of 13,500 to 22,000 x 10^6 t yr⁻¹.

Several of the rivers in Table 2, however, are severely affected by human activity. The most striking example is the Nile. The measured mean annual sediment load during the years 1902–63 was 160–178 x 10⁶ t yr⁻¹. After the construction of the Aswan dam in 1964, no sediment passed downstream. As a consequence, the delta receded and the delta rim is now situated about 5 km behind its most advanced position (Khafagy and Fanos 1993). The Mississippi is another example of a river where the sediment load has decreased because of reservoir sedimentation. The many reservoirs built on its tributaries reduced the transport from 400–500 x 10⁶ t yr⁻¹ to the present load of 230 x 10⁶ t yr⁻¹ given in Table 2.

Syvitski et al. (2005) estimated that 20% of the global load is deposited in reservoirs that were mainly constructed within the past 50 years. Sediment transport in the Yellow River has decreased because of reduced precipitation, increased water abstraction and sediment-control practices. During the years 1950–77, the river had a load of 1.6 x 10⁹ t yr⁻¹ (Milliman and Syvitski 1992). According to the Yellow River Commission, the mean transport rate in its lower part had decreased to 0.5 x 10⁹ t yr⁻¹ for the period 1989–2003.

Some of the rivers draining to the Arctic Ocean have also experienced large sediment-transport changes due to dam construction and other human impacts. Hasholt et al. (2005) gave a recent estimate of the total sediment transport to the Arctic Ocean and adjoining cold seas. By combining available monitoring data and estimates for ungauged areas, they estimated that the total sediment transport is within the range $325-885.1 \times 10^6$ t yr⁻¹. Of this total, only a part can be considered as monitored, while the rest is based on different types of estimate, ranging from morphology- and processbased estimates to more empirically based estimates. The largest uncertainty is the contribution from glacier calving, which may be as large as 500×10^6 t yr⁻¹.

The sum of the load carried by Russian rivers is also of considerable magnitude, with a total of 73×10^6 t yr⁻¹. The water discharge of the Yenisey river ranks among the six largest in the world, averaging an annual total of 630 km³ into the Arctic Ocean. Its sediment load is, however, relatively low. During the period 1941–56, the mean sediment load was measured at 13.2



Figure 2. Geochemical distribution of lead in overbank-sediment samples from 747 localities in Europe. After Salminen et al. 2005.

x 10⁶ t yr⁻¹ at the Igarka gauging station in the lower part of the river basin. After dams were constructed on the Yenesey and its tributary the Angara, the sediment load decreased to a mean of 4.2 x 10⁶ t yr⁻¹ (Bobrovitskaya et al. 1996). Observations of sediment transport in other Russian rivers draining to the Arctic Ocean are, according to Holmes et al. (2002): Lena 20.7 x 10⁶ t yr⁻¹ (7.6–40.0 x 10⁶ t yr⁻¹), Ob 15.5 x 10⁶ t yr⁻¹ (13.0–16.6 x 10⁶ t yr⁻¹), Kolyma 10.1 x 10⁶ t yr⁻¹ (4.7–16.1 x 10⁶ t yr⁻¹), Pechora 9.4 x 10⁶ t yr⁻¹ (6.5–13.5 x 10⁶ t yr⁻¹) and Severnaya Dvina 4.1 x 10⁶ t yr⁻¹ (2.5–6.6 x 10⁶ t yr⁻¹).

The long-term sediment-transport monitoring programme of the river Kolyma in east Siberia, is a striking example where human impacts have increased the sediment load. Part of the long-term record of water discharge and sediment load initiated in 1941 is shown in Figure 3. Although the mean annual water discharge has remained fairly constant, there has been a gradual increase in sediment yield since 1965. Among the anthropogenic factors affecting the sediment yield, gold mining is important because the surface soils are removed over large areas, thus producing conditions suitable for intensive erosion (Bobrovitskaya 1996).

The Kolyma case illustrates the way that analyses of overbank sediments may be used to obtain information of the present and past fluxes of geochemical elements. The upper sample represents the present conditions; dating the sedimentary sequence would help identify older overbank deposits representative of conditions

Table 2. The suspended-sediment flux of a selection of major rivers compared to the global total.

River	Area 10 ³ km ²	Sed. load 10 ⁶ t yr ⁻¹
Amazon	7180	363
Mississippi	3221	230
Parana	890	90
Colorado	629	135
Congo	3822	65
Niger	430	40
Nile	2881	178
Yangtze	1980	486
Yellow river	745	1600
Indus	960	100
Ganges/Bhramaputra	1480	1670
Mekong	783	150
Irrawaddy	431	170
Flux to Arctic Ocean by major rivers, excluding calving		384
Sum		5661
Global total		13500

before modification by human impact. Since overbank sediments are deposited during major floods and represent conditions during which the main volume of sediments are transported, it may be possible to detect the long-term geochemical changes in the sediment load that is delivered to the oceans. The deltas of large rivers are often complex features, in which the sediments accumulated at the ocean margin throughout long periods. It may thus be easier to recognise the age of older sediments if the delta history is known at least as far back as the time when pristine conditions prevailed, perhaps 200 years before present or more.

The need for a global project

Present data on the chemical composition of the Earth's surface is limited to small, detailed studies in restricted areas. It is, however, necessary to map the global distribution of chemical elements to reveal large-scale patterns, both natural and anthropogenic. If one continued to use the present techniques and approach it would probably take several hundred years to complete the task. A wide-spaced sampling programme of overbank



Figure 3. Sediment load and runoff in the Kolyma river in the period 1941–89. After Bobrovitskaya 1996.

sediments of rivers is the only practical way to obtain a global overview in a short time.

Such an overview is of practical importance in exploration for mineral resources. China has devised a mineral exploration strategy to rapidly assess the mineral-resource potential of a very large area by wide-spaced geochemical mapping followed by a progressive reduction of target areas by denser sampling (Xie and Hangxin 2001). The geochemical distribution of platinum in China was determined by analysing only 529 overbanksediment samples. The most promising Pt target is a huge geochemical megaprovince covering an area of 700,000 km² in southwest China. Many Chinese exploration parties and foreign exploration companies have now commenced exploration activities. It is believed that similar megaprovinces will be found through a global mapping project.

In existing mining areas where river sediments are heavily polluted, it has previously been difficult to identify new mineral deposits. The application of pre-mining overbank-sediment data can successfully be used for prospecting in such areas.

Wide-spaced floodplain sampling is also a useful tool for environmental monitoring purposes, as demonstrated by the resultant mapped distribution of the ratio of Hg content in surface overbank sediments (present time) to Hg content in deep overbank sediments (historical time). It is obvious that Hg pollution in eastern China is serious (Xie and Hangxin 2001).

Other investigations have revealed that 30% of the floodplains in Europe are polluted, mainly by mine waste (Bølviken et al. 1996). A Pb map for overbank sediments in Europe is shown in Figure 2. Northern Germany, Belgium and England are particularly heavily polluted (Salminen et al. 2005).

A high natural content of a poisonous element can also represent a threat. An example is the high arsenic content of floodplain sediments in India and Bangladesh. Recent findings show that similar conditions may also exist elsewhere. This naturally high As content has resulted in hazardously high concentrations of arsenic in the groundwater used for drinking. Six million people in west Bengal use water with an unsafe arsenic level. Global geochemical mapping will reveal areas where the chemical quality of the groundwater should be investigated further. Rivers are large transporting systems that continuously redistribute and disperse the sediments in their catchments. Systematic mapping based on overbank-sediment samples may be used to identify source sediments and prevent further erosion of polluted deposits.

It is of great importance to know how future climate change will affect river systems. The globally averaged surface temperature is projected to increase by 1.4 to 5.8°C over the period 1990 to 2100. The water vapour concentration and precipitation are also projected to increase (World Meteorological Organization 2007). A probable result is that some permafrost will melt and more sediments will be released into river systems.

In Norway, for instance, model simulations carried out by RegClim (2005) predict an increase in total annual precipitation towards the year 2100. The increase will vary between 5% and

20% in different parts of the country and extreme rainstorm frequency will also increase.

Extreme floods have often been observed to cause erosion in deposits that are not exposed during floods of lower recurrence intervals. The 1995 flood in Norway was a large-magnitude flood of 100–200 year recurrence interval. During this event, the concentrations of particle-bound metals Cu, Zn, Cd and Pb increased considerably at a downstream monitoring station where the large river Glomma enters the sea. This increase in metal content is believed to be due to extensive erosion in the mining areas in the upper part of the catchment (Holtan and Holtan 1996, Bogen and Bønsnes 2000). The increased frequency of extreme situations will become a global phenomenon. Largescale changes in water discharge and sediment load will in the long term affect the chemical composition of the oceans. Changes in sediment supply not only cause costal retreat, but also greatly affect the benthic environment and coastal fisheries.

Conclusions

This paper discusses the use of overbank sediments from floodplains as a sampling medium to obtain large-scale geochemical maps. Low-density geochemical maps based on overbank sediments have been shown to give reproducible patterns that coincide with the patterns of high-density maps of the same areas.

Traditional prospecting methods based on active stream sediments involve sample collection without the use of skilled personnel. The appliance of overbank sediment samples is more costeffective, but requires knowledge of processes of erosion, transport and sedimentation within river basins. Such information adds another dimension to geochemical mapping. Knowledge of sedimentary dynamics makes it possible to reveal the historical development of a river system and record the associated changes in its chemistry.

Depth-integrated samples of overbank-sediments reflect the composition of many current and past sediment sources upstream of the sampling point, in contrast to active stream sediments which normally are recent deposits derived from a more restricted number of actively contributing sediment sources. In many regions, overbank sediments are more representative of drainage basins than active stream sediments and, consequently, can be used to disclose geochemical distribution patterns on a regional to continental scale by means of widely scattered sampling at low cost per unit area.

The stratigraphy of overbank sediments may in some cases be complicated. However, in flood plains or old terraces along laterally stable or slowly migrating channels, it is normally possible to obtain recent sediments near the surface and preindustrial sediments at depth. Mapping the composition of recent and pre-industrial overbank sediments can, therefore, be used (1) in a characterisation of variations in the natural geochemical background as well as a documentation of the present state of pollution, and (2) as a regional prospecting tool in natural as well as polluted environments.

Possible vertical movements of elements in overbank sediment strata have been reported, especially in some studies of relatively mobile elements (arsenic and cadmium) in noncalcareous areas heavily influenced by acid rain. However, the overall impression is that such migration is not a major problem in the use of overbank sediments in geochemical mapping.

Human interference with rivers and the predicted climate change will affect the global sediment flux to the oceans. Investigations of sediment sources and past changes in the particle-associated transport of chemical elements, documented by the sedimentary record, will be a valuable tool, which can contribute to a better understanding of future developments.

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